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IR WINDOW STUDIES

John H. Marburger

University of Southern California

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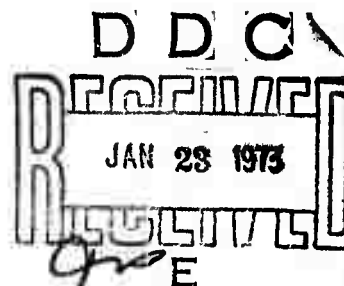
UNIVERSITY OF SOUTHERN CALIFORNIA

IR WINDOW STUDIES

John H. Marburger

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For the Period Ending 31 August 1972

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Solid State Sciences Laboratory

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13. ABSTRACT The initiation of a joint theoretical and experimental program on the preparation, characterization, and evaluation of IR Window Materials is reported. During this first quarter of operation, most of the activity has been design and fabrication of facilities for the program. Preliminary results regarding the characterization of point defects in CdS and CdTe by tracer self diffusion and high temperature Hall mobility techniques have been obtained. Other areas in which preliminary investigations have been carried out include closed tube chemical vapor growth of GaAs samples, measurements of thermal shock and ductility of alkali halide samples, and theoretical analyses of IR absorption mechanisms. We also report the completion of a general two dimensional computer code for the analysis of optical properties of IR windows.			

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ROLL

WT

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Alkali Halides

III-V Semiconductors

II-VI Semiconductors

Thermal lensing

1b

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CONTENTS

	Page
ABSTRACT	1
1. INTRODUCTION	
1.1 General Objectives	3
1.2 Work Statement and Project Identification	4
2. PROGRESS BY PROJECT	
a.1 Effect of Oxygen and Other Impurities on I. R. Absorption in II-VI and III-V Compounds	7
a.2 Optimization of Alkali Halide Window Materials	8
a.3 Growth of Crystals for IR Window Research	13
b.1 Fabrication of Polycrystalline IR Window Materials	16
c.1 Mechanical Behavior of III-V and II-VI Compounds	19
d.1 Surface and Interface IR Absorption	20
d.2 Study of Defects in II-VI Compounds	23
e.1 Theoretical Studies of Absorption Mechanisms in IR Window Materials	27
f.1 Techniques for Indirect Measurement of Small Absorptive Losses	30
g.1 Characterization of Optical Performance of IR Window Systems	32
3. DISCUSSION	44
4. SUMMARY	45

1. INTRODUCTION

1.1. General Objectives

The infrared window program at the University of Southern California is a cooperative effort involving faculty from the departments of Electrical Engineering, Materials Science, and Physics. Each of the projects in this program falls under one of the following general categories

- A. Materials growth and window fabrication
- B. Materials characterization
- C. Materials evaluation for window applications.

Projects in category A include the preparation of crystals of alkali halides, GaAs and other promising semiconductor materials; liquid epitaxial growth of very high purity GaAs and hot pressing of semiconducting compounds for window fabrication. Category B includes mechanical characterization of semiconductors and alkali halides, defect characterization through absorption spectroscopy, tracer self diffusion, high temperature Hall effect measurements, and X ray and electron microprobe techniques when relevant. In this category also falls studies of surface absorption and conductivity, and theoretical analysis of IR absorption mechanisms. Category C includes calorimetric determination of absorption coefficients, investigations of possibly more sensitive absorbance measuring techniques, mechanical testing of windows, and numerical simulation of optical and thermal window performance.

The general objectives in the initial phase of of the program are:

1. Establish a local facility for calorimetric determination of small IR absorbances using a powerful CO_2 laser tunable from 9 to 11 μm .
2. Establish facilities for the preparation of materials used in the program.
3. Establish a basis for theoretical support of the program objectives.
4. Redirect continuing USC programs (some supported by other contracts in the past) toward objectives relevant to the IR window problems.

- c. "Investigate the mechanical properties of different window candidate materials and their modification by different processing techniques."
- c.1 Mechanical Behavior of III-V and II-VI Compounds
S. M. Copley
(a.2 also includes mechanical testing)
- d. "Characterize the candidate window materials by chemical, physical, metallurgical and other processes. This includes materials obtained from other sources as well as those prepared under subparagraph a. above. Particular emphasis will be placed on the understanding and control of the mechanisms by which chemical and physical defects modify the properties of the materials."
- d.1 Surface and Interface IR Absorption
C. R. Crowell, J. M. Whelan
- d.2 Study of Defects in II-VI Compounds
F. A. Kröger, M. Gershenzon
- d.3 Determination of Maximum Acceptable Impurity Concentrations in IR Window Materials
M. Gershenzon
- e. "Conduct theoretical investigations to enhance the understanding of the basic mechanisms of absorption of optical energy, particularly but not restricted to, the region around 10.6 micrometers wavelength. Emphasis will be on those mechanisms which may dominate the residual absorption, i. e. the very low loss materials (for example, potassium chloride)."
- e.1 Theoretical Studies of Absorption Mechanisms in IR Window Materials
R. W. Hellwarth
- f. "Investigate and improve methods of measuring the optical absorption of materials which have very low losses at 10.6 micrometers in order to correlate theoretical and experimental data on candidate window materials."
- f.1 Techniques for Indirect Measurement of Small Absorptive Losses
W. H. Steier

g. "Evaluate the optical performance of candidate window materials. This investigation shall include detailed studies of the influence of physical material parameters on degradation of the optical performance of the window materials (for example, thermal lensing). The study of promising window designs, such as multiple component windows and material criteria for these designs, will be considered."

g.1 Characterization of Optical Performance of IR Window Systems

J. H. Marburger

If additional projects are added during the course of the contract, they will be assigned codes according to this scheme.

2. PROGRESS BY PROJECT

a. 1. Effect of Oxygen and Other Impurities on IR Absorption in II-VI and III-V Compounds

J.M. Whelan, M. Gershenzon

A system has been designed for growing high purity selectively doped GaAs films to be used in surface and bulk defect absorption studies. It is based on low temperature solution growth of thin films and can be operated in a temperature gradient made to grow relatively thick films. Features of this system include a horizontal sliding boat-substrate holder arrangement for limiting the effective solution volume. Variations of this have been used for some time. A much less common feature will be the use of a specially designed furnace in which the longitudinal temperature gradients are minimized without giving up the provision for temperature control to within $\pm 0.02-0.05^{\circ}\text{C}$. Materials for this system are on order and the initial fabrication has begun.

a.2. Optimization of Alkali Halide Window Materials

P. J. Shlichta

The objectives of the first phase of this program are:

- (1) Growth of high-purity and doped NaCl and KCl crystals.
- (2) Testing to determine the effect of each impurity or defect on:
 - (a) Optical absorption at 10.4 microns
 - (b) Thermal shock resistance
 - (c) Yield stress
 - (d) Brittle-ductile transition temperature

Thus far the following activities have been initiated:

Purification of NaCl and KCl: On the basis of a recent review of purification techniques (ref.1), it was decided that both ion-exchange and gas treatment (i.e., with HCl and Cl₂) would be necessary. Moreover, it was decided that, for reasons of speed and reliability, it was preferable to undertake these activities at USC rather than rely on an outside contractor. The proposed purification scheme is outlined in Figure 1. This scheme provides us with several options with respect to controversial aspects of purification:

- (1) It has been reported (ref.2) that treatment of the molten salt with CCl₄ vapor removes anionic impurities much more rapidly and completely than HCl treatment. However, this procedure may leave behind impurities such as CO₃⁼ and CO₂Cl⁼.
- (2) Treatment with Cl₂ may be insufficient to burn off traces of organic material, in which case it may be necessary to follow Grundig's procedure (ref. 3) of treating the solid salt with 10 mm of O₂ at 500°C.
- (3) It is uncertain whether the purified salt can be melted in fused silica without introducing silicate as an impurity. Accordingly the apparatus has been designed to provide alternatives for each of the above steps. Thus far, all the necessary equipment has been ordered and it is hoped that the system will be operational by the end of the year.

Crystal Growth: A Lepel crystal puller is being modified so as to eliminate all metal parts from the growth chamber, e.g. the seed holder and pull rod are being replaced by carbon parts and new polycarbonate end plates

are being made. This apparatus should be operational by the end of the year. In addition, an auxiliary puller will be mounted onto the gas treatment apparatus so that crystals for analytical and optical measurements can be grown during the purification process.

Thermal Shock: Preliminary tests of thermal-shock resistance were made by impinging a 2 mm oxygen-propane torch flame on the center of 20 x 20 x 2 mm NaCl plates for five seconds. When the plates were initially at room temperature, the thermal shock invariably initiated cleavage cracks. However, when the crystals were preheated to 300°C (i.e., well above the brittle ductile transition temperature), no cracking resulted, even when the flame was allowed to melt a bead on the face of the crystal. Further experiments showed that preheated crystals could even be welded together by the gas torch.

In the next quarter, these tests will be continued using a focused laser beam in place of the gas flame.

Ductility: According to the literature, (refs. 4, 5), there exists a brittle-ductile transition temperature below which, alkali halide crystals are ductile in certain modes of deformation (such as [100] tension or compression and [100]/[001] bending) but brittle in others (such as [111] tension or compression or [100] torsion). Above the transition temperature, the crystals are completely ductile.

To confirm these observations, a series of qualitative tests were made using 5 x 5 x 75 mm commercial NaCl cleavage rods. The results were as follows:

	[100]/[001] BENDING	[100] TORSION
ROOM TEMP. (AS-RECEIVED)	BRITTLE	BRITTLE
ROOM TEMP. (WATER POLISHED)	DUCTILE (~10%)	BRITTLE
350°C. (AS-RECEIVED)	VERY DUCTILE (>30%)	DUCTILE (>15%)

It appears that heating the crystal above the transition temperature not only activates new slip systems but also eliminates the embrittling effects of surface damage and/or interaction with the atmosphere.

With respect to experiment design, the data above suggest that [100] torsion in a thermal gradient will provide the quickest method of determining the approximate transition temperature for a given specimen. Such an apparatus is being constructed and should be operational sometime during the next quarter. More precise measurements of the yield stress above and below the transition temperature will probably be made by [111] and [100] compression tests.

Application to Laser Windows: The most likely failure modes of a CO₂ laser window are (a) cracking, (b) sagging or other plastic deformation, (c) differential heating and consequent optical "blooming", and (d) excessive light scattering. Of these, (a) would appear to be the most catastrophic since it can destroy the entire system. This, apparently, can be avoided by heating the entire window so that all parts of it are above the transition temperature; the thermal shock experiments reported above seem to confirm this concept. Unfortunately, this remedy for cracking will almost certainly increase the tendency of sagging, the critical parameter being the yield stress just above the transition temperature. According to this approach, window performance might be optimized by (1) purifying the material so as to minimize the transition temperature, and (2) finding an impurity or defect which raises the yield stress but does not raise the transition temperature or the infrared absorption.

An alternative approach would be to use "forged" polycrystalline windows. Because of latent hardening (ref. 6), the yield stress would be almost a magnitude higher. However, unless the grain size is exceedingly small, the tendency toward cracking would be comparable to that of single crystals; therefore, such windows would probably also have to be operated above the transition temperature. Even in this region, however, they would have several times the yield stress of single crystals. (ref. 4) The use of such windows may, however, be limited by their tendency to excessive light scattering.

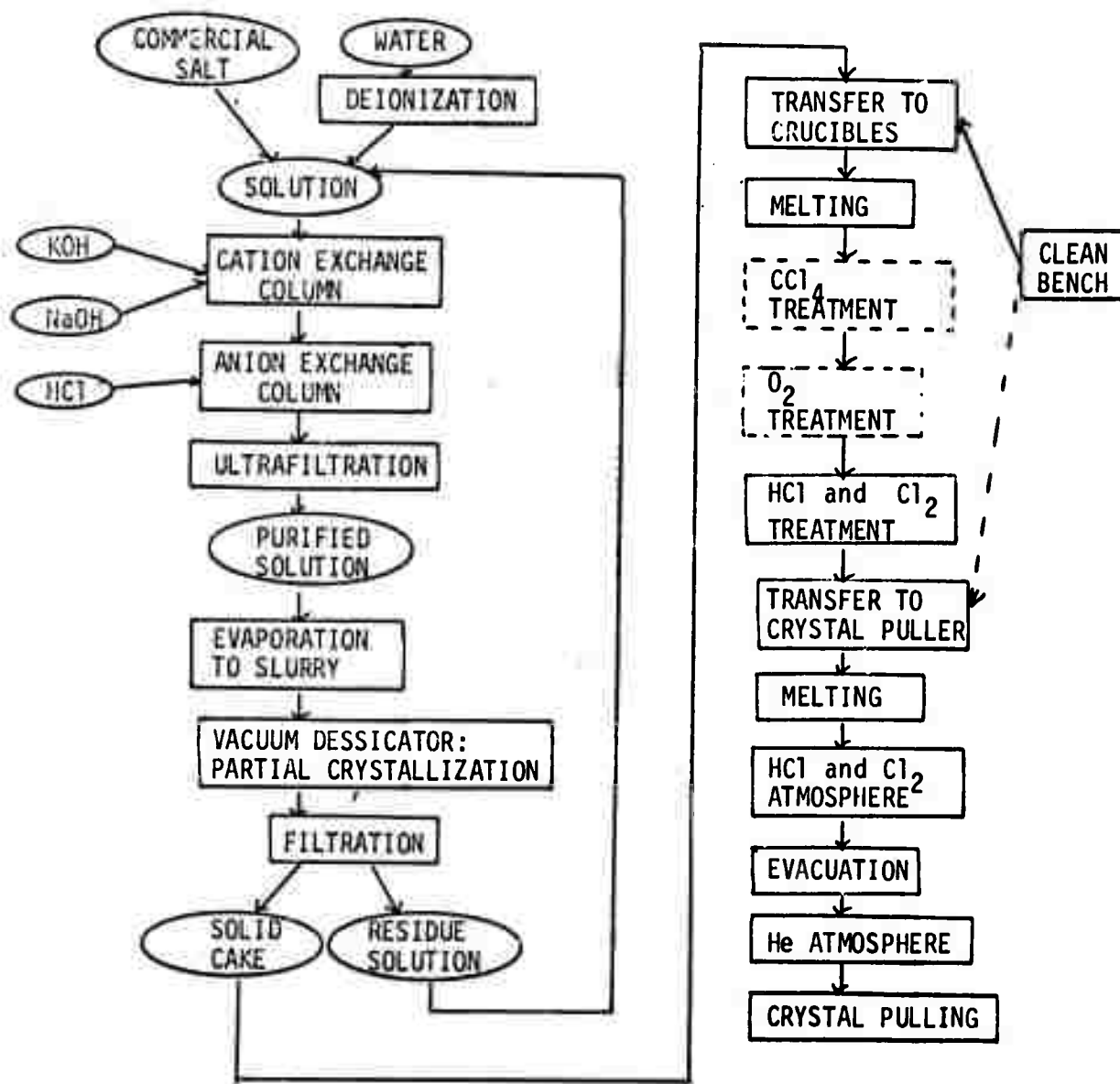


FIGURE L. Operational Diagram of Purification System for NaCl and KCl.

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a. 3. Growth of Crystals for IR Window Research

W. R. Wilcox

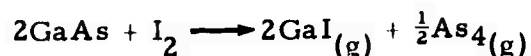
(Chemical Vapor Deposition of Gallium Arsenide)

1. Purpose

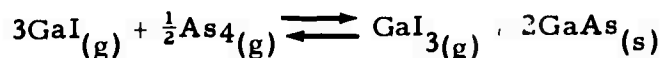
This study is being undertaken in an attempt to produce high purity GaAs as needed for IR laser window research. The major advantage of a C.V.D. process over other methods of GaAs growth, is that growth can take place at lower temperatures, thus minimizing the problems of impurity solubility at high temperatures and crucible contamination.

2. Basic Reactions

The basic chemical reaction in the CVD process involves the transport of Ga as either a volatile halide, oxide, or alkyl compound from a hot to a cold zone of a reaction tube. For example in our preliminary work, an amount of I_2 or HCl (to give ≈ 1 atm. pressure at 900°C) was added to an evacuated tube containing GaAs. The tube was subsequently sealed. The initial reaction between GaAs and I_2 is:



GaI now reacts with arsenic vapor forming GaI_3 and GaAs according to:



Transport may be obtained from a hot to a cold area since this equilibrium favors GaAs deposition in the cold zone.

3. Experimental Work

When nucleation occurs in the tip of a capillary, there is an increased chance that as growth proceeds up the capillary, one crystallite may dominate the growth pattern. Therefore various tube geometries and methods of applying a cold spot were tested. Several tests were conducted using capillary tips and forced air cooling to induce crystallization in the tip. Also the tubes were placed in a furnace with a $50\text{-}100^\circ\text{C}$ temperature difference between the feed and seed

zone. In a few cases promising results were obtained. Although the crystals grown were polycrystalline, there were only small voids in them. In addition the growing surface was convex with respect to the solid. This will tend to produce minimal dislocations since dislocations tend to propagate normal to the growing interface, and in this case will tend to grow out of the crystal. In a typical example (Run #8) a polycrystalline sample is grown by the HCl transport method using a copper wire cemented to the end of the tube as a heat sink. The temperature of the feed and seed zone were 880 and 820°C respectively. The crystal was cut and mechanically polished, and then submitted to Dr. Steier's group for IR absorption measurements by calorimetric methods.

In some of the preceding experiments, it was observed that nucleation of individual crystallites occurred on the sides of the capillary along with nucleation in the tip. This could be due to insufficient temperature difference between the cold spot in the tip and the walls, especially as the growing crystal advances.

An alternative that may alleviate this problem is the travelling heater method. A sealed tube containing GaAs + I₂ is drawn through a sharp temperature gradient at a slow rate of approx. 1 mm per day. In this way the growing crystal can establish a steady state growth rate vs. distance profile with respect to the temperature gradient. Two travelling heater furnaces were constructed and preliminary runs indicate the need for a small vapor space for transport. This small vapor space is also needed to minimize the effect of the vapor phase analog of constitutional supercooling. This can be a problem, because of concentration gradients caused by diffusion or convection, the partial pressures of As₄ + GaI may be greater than the equilibrium partial pressures at the particular growth site.

We also found that it was necessary to obtain GaAs starting material free from excess gallium. It was observed in several cases with

excess Ga in the starting material, no crystal growth would take place, only Ga transport occurred. This will create some experimental problems in sealing quartz tubes with a small vapor space because of thermal decomposition of GaAs.

4. Future Work

Further experiments with various tube geometries are planned. One particular tube design consists of an internal projection with a small flat surface cooled by a stream of air. An advantage of such a design is that crystal growth from such a projection would be free of constraints from the walls of the tube.

Several more travelling heater furnaces are to be placed in operation in order to investigate this method of growth in a reasonable period of time. In addition, further work is planned for stationary tube methods, mainly in controlling the temperature profile at the seed end, and testing different capillary sizes and shapes.

5. Open Tube CVD Facility

A system has been designed and parts ordered for a CVD open tube system that will allow one to investigate several transport techniques. It is planned to investigate Ga transport by volatile HCl, HI, and H₂O vapor compounds.

Special attention will be directed toward investigation of CVD processes using alkyl gallium compounds and arsine as transporting agents. Thick films (≈ 1 mm thick) have been grown at very fast rates (≈ 1 mm/day) using this method. Since arsine is highly toxic, the whole system will be installed in a special hood which is now under construction.

b. 1. Fabrication of Polycrystalline IR Window Materials

S.M. Copley, J.M. Whelan

The goal of this portion of the program is to produce samples of polycrystalline gallium arsenide by a novel hot pressing technique involving the use of volatile sintering aids. Working drawings of the special hot press have been prepared and materials for its fabrication have been ordered.

In our technique, excess arsenic will be employed to enhance the sintering of gallium arsenide powder. In addition to the "lubricating" properties of molten arsenic which will enhance particle rearrangement during sintering, the high volatility of arsenic and its oxides will be helpful in removing the excess lubricating arsenic and contaminating oxides from the pressed compact. However, the high vapor pressure of molten arsenic (28 atmospheres) presents a containment problem. A schematic diagram of the hotpress is shown in Fig. 1. It is planned to seal the arsenic vapor in the hotpress die using a molten glass which will be retained in a well on top of the die by pressurizing the hotpress chamber with argon.

The following operating procedure is envisioned. First the die (loaded with gallium arsenide powder, and containing chips of glass in the recess on top) will be installed in the furnace chamber which will then be evacuated. A load will then be applied to the die plunger by pressurizing the diaphragm cylinder. Then, while evacuation continues, the furnace will be heated to melt the glass with the chill operating to keep the excess arsenic cool (below 250°C). When the glass is molten, the vessel will be pressurized with argon, the chill will be turned off, and the furnace temperature will be adjusted to the desired hotpressing value.

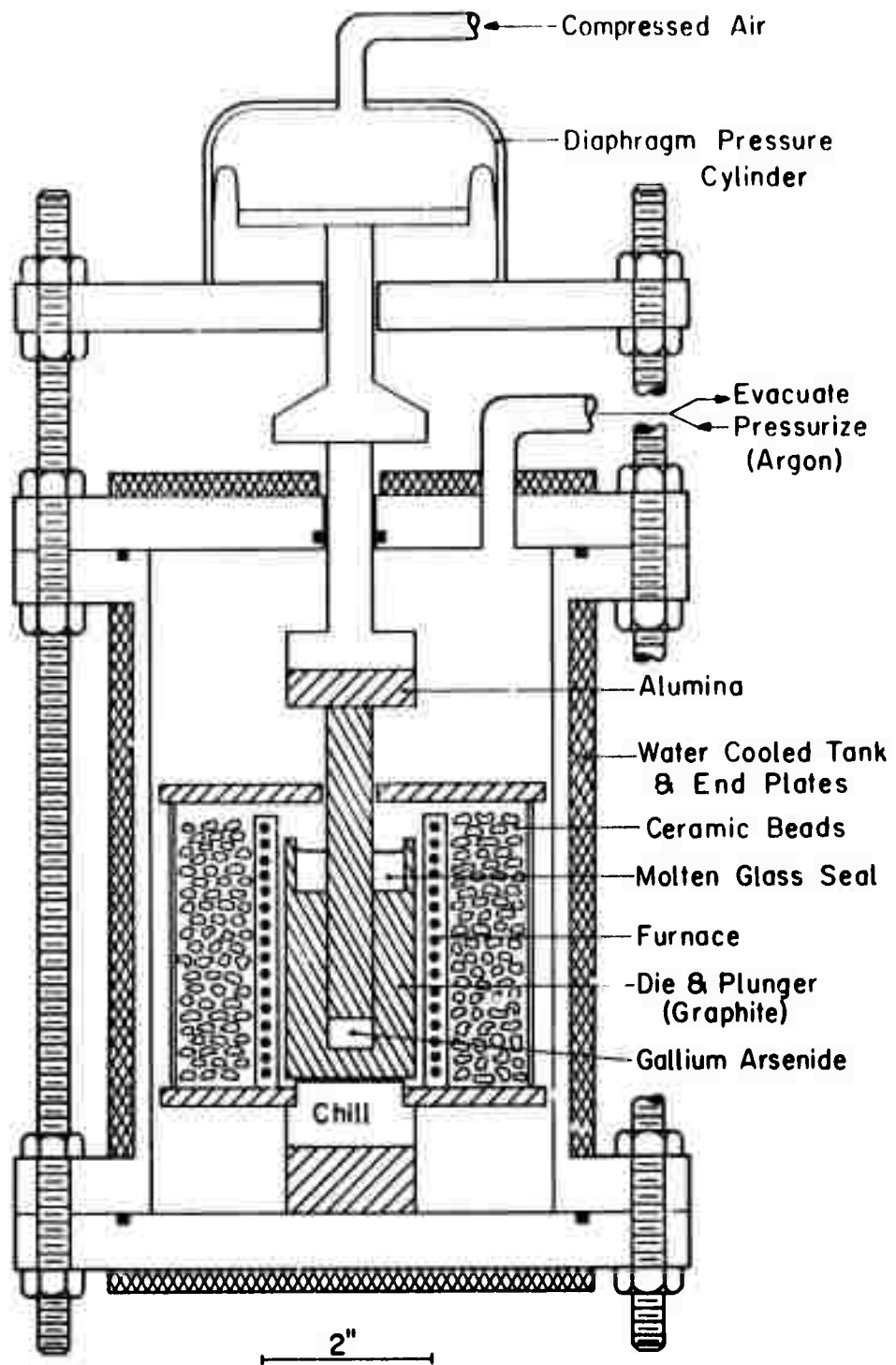
After compaction the furnace will be allowed to cool, and the applied load and argon pressure will be removed. Gallium arsenide has an unusually low coefficient of thermal expansion ($6 \times 10^{-6}/^{\circ}\text{C}$) and the

graphite die may contract around the pressed compact during cooling. The use of expendable dies has been considered.

After cooling, the arsenic rich compact is removed from the die. Post curing of the compact for final adjustment of arsenic content is being considered.

Production is planned for gallium arsenide dies 1/2" diameter by 1/8" thick. Hotpressing capabilities to 5000 psi (on 1/2" diameter ram) and 1200°C are planned.

During the next quarterly period the hot press will be fabricated at the USC Engineering Machine Shop.



Hotpress Utilizing Volatile Sintering Aids

c.1. Mechanical Behavior of III-V and II-VI Compounds

S. M. Copley

This investigation is concerned with a determination of the mechanical behavior of various window candidate materials. A study of the mechanical behavior of single crystals of GaAs doped with Si has been initiated.

A 350 gm melt grown single crystal of Si-doped GaAs with a carrier concentration of $1.8 \times 10^{18} / \text{cm}^3$, a mobility of $1900 \text{ cm}^2 / \text{v. sec}$ and a resistivity of $1.89 \times 10^{-3} \Omega \text{ cm}$ was obtained from Crystal Specialties, Inc.* Compression specimens with length to width ratios of 1, 1.5 and 3.0 were prepared by a series of operations including initial shaping with a precision cut-off wheel, two-circle goniometer unit to give selected orientations and precision grinding. Specimens with $\langle 100 \rangle$ and $\langle 111 \rangle$ stress axes were selected for the initial stress-strain experiments on the basis of Schmid factor considerations. The $\langle 100 \rangle$ stress axis favors slip on $\{111\} \langle 1\bar{1}0 \rangle$, the previously observed slip system, while the $\langle 111 \rangle$ stress axis favors slip on other systems.

A graduate student has been trained to carry out the stress-strain experiments on the CGS Lawrence Mechanical Testing System. In the next quarterly report, stress strain data should be available at various temperatures. This data will indicate the operative slip modes in GaAs single crystals and the temperature dependence of the critical resolved shear stress for these modes. It will also indicate the effect of varying the length over width ratio of the specimens on stress-strain results.

*Crystal Specialties, Inc.
419 W. Maple
Monrovia, California 91016

d. 1 Surface and Interface IR Absorption Under Intense Illumination
C. R. Crowell, & J. M. Whelan

We have undertaken to attempt to find evidence for surface and interface IR absorption through electrical measurement of conductivity and interface charge in GaAs MOS type structures with air gap insulation and with typical antireflection coatings. During this initial period we have taken preliminary steps toward establishing a sample geometry.

To study the air-GaAs interface we have decided on the geometry which will be described below in detail.

Since most antireflection coatings used for IR laser windows can be deposited fairly easily on semiconductors, a typical structure will be used for antireflection coating-GaAs interface absorption studies.

Structure for the Study of Air-GaAs Interface: (Figure 1)

For this structure, gate electrodes are evaporated on optically flat substrate and Schottky barrier spacitors are deposited on freshly prepared flat GaAs surfaces. Evaporated ohmic contacts are used for ohmic contacts on GaAs and present techniques for making ohmic contacts appear adequate.

For sensitivity in investigating surface states with an air dielectric structure the large difference in dielectric constant between the air and the GaAs demands that the air gap thickness be as small as current fabrication techniques make possible. We are initially aiming at $\leq 2000\text{\AA}$. This places importance on the flatness of GaAs surface and gate electrodes.

We have attempted to establish the thickness of the air gap by the thickness of the evaporated metal film on the GaAs. The film is deposited in a pattern determined by a shadow mask which was designed and fabricated during this reporting period. During device operation this film will be contacted by the metallized registered pattern deposited

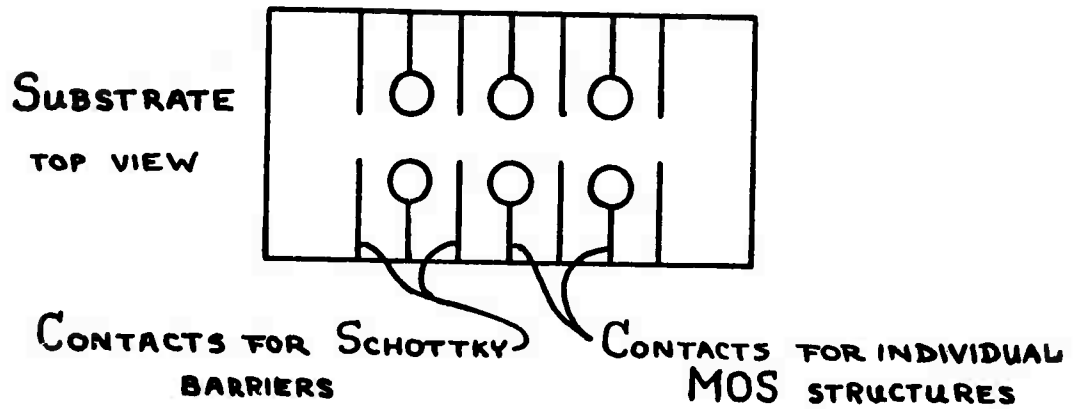
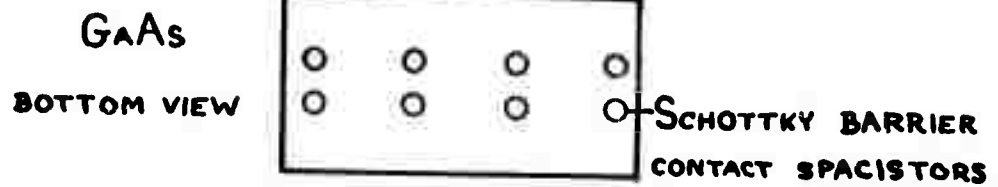


FIGURE 1a.

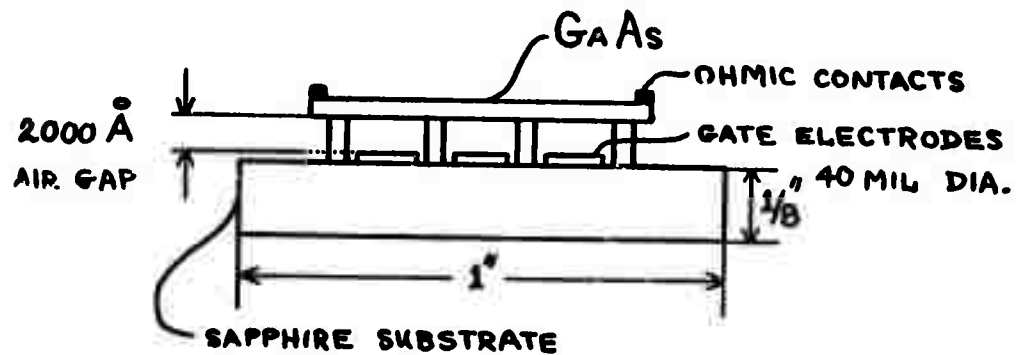


FIGURE 1b.

on the insulating substrate which is also used to support the gate electrode. The registering pattern and metal gate is deposited simultaneously. Thus we have both MOS and Schottky barrier electrodes to compare the behavior of the GaAs bulk and the sum of surface and bulk effects. Considering the order of thickness dimensions, for dimensional integrity we would like to stay away from photoresist techniques.

We have also initiated studies to improve the flatness of our polished GaAs samples. A preliminary examination of chemically-mechanically polished GaAs by multiple beam interferometry suggested that flatness within 2000\AA could be achieved relatively easily over a 40 mil diameter area. This should be satisfactory for MOS-type studies but may not be adequate for a MOSFET stripe geometry which we also plan to use in our experiments.

During the next quarter, we will continue our work on preparing flat GaAs samples and begin work on the appropriate circuitry for pilot electrical measurements.

d. 2. Study of Defects in II-VI Compounds

F. A. Kroger, M. Gershenzon, S. S. Chern, H. R. Vydyanath

The long wavelength optical absorption of 2-6 compounds depends to a large extent on the presence of point defects, atomic defects such as vacancies and interstitials as well as free electrons or holes. Minimalization of absorption near 10.6μ as required for an ir window requires minimalization of the defects giving rise to absorption in that range. This can be done only if we know what types of defects are present and how their concentration depends on the conditions of preparation, and what is the absorption due to each defect. The latter is related to the position of the energy levels of the defect in the forbidden gap.

Determination of the defects present and the ways in which their concentration varies with the conditions of preparation is one of the goals we have set ourselves. Methods to achieve this goal include measurements of self diffusion of the components and the Hall effect, each as a function of temperature and the component activities. Earlier work on CdS has shown that the following species are involved: $V_S^{\cdot\cdot}$, Cd_i^{\cdot} , V_{Cd}'' , S_i^x , e' , h' . We expect similar species in CdTe. This is born out by evidence available so far; the only difference being that $Cd_i^{\cdot\cdot}$ appears instead of Cd_i^{\cdot} (and Te_i^x instead of S_i^x). Since free carriers are known to absorb in the infra red, steps have to be taken to minimize the concentration of electrons and holes. For CdS this can be done by heating at a high sulfur pressure and/or by doping with acceptors. Silver may be a suitable dopant. For CdTe, in which both n- and p-type conduction has been observed, it is necessary to stabilize the Fermi level near the middle of the gap. This has been found to be difficult if not impossible in undoped material, but is possible upon weak doping with donors combined with preparation in a tellurium atmosphere. Under these conditions, the donor (In, Cl) is compensated by native defects (V_{Cd}'' or S_i'')

level 0.05 eV below the conduction band is probably also due to one of these defects (the A_2'' center). In any case, it must be the one present as a minority. If foreign donors such as In are present, associates $(\text{In}_{\text{Cd}}\text{V}_{\text{Cd}})'$ or $(\text{In}_{\text{Cd}}\text{Te}_i)'$ may also be present, and the levels caused by these species have to be considered. A possible level scheme for CdTe is shown in Fig. 1. At any level involving Te_i or V_{Cd} , these symbols may be interchanged, V_{Cd} levels being Te_i levels, and vice versa, the energy separation corresponding to a wavelength of 10.6μ is also indicated. For a good window material all levels having a separation from the bands of this order or less must be empty (when close to the conduction band) or occupied (when close to the valence band): the Fermi level should be more than 0.2 eV from the band edges. Experiments carried out under the contract and other ones intended to be carried out in the next period are listed below.

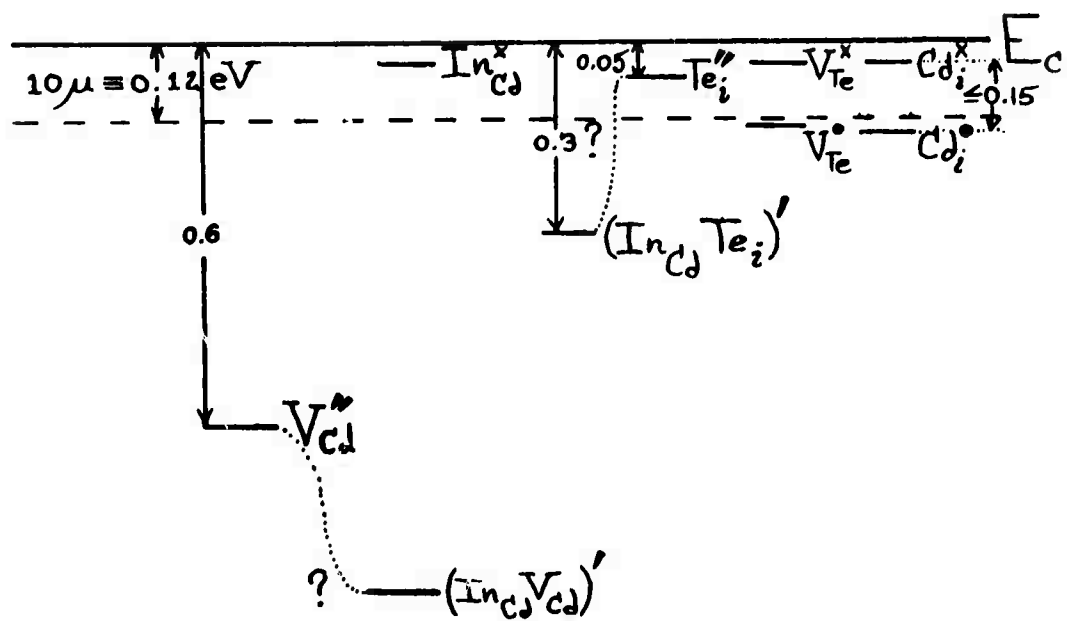
1. Concentration profiles of Indium diffusing into CdS and CdTe at 800°C show an abnormal S shape, indicating a diffusion coefficient varying from medium at the surface, to large $\approx 200 \mu$ below the surface, to small deeper into the crystal. The reason for this behavior is not clear.
2. Te^* tracer self diffusion in CdTe at low p_{Cd} is known to occur by neutral interstitials, $D_{\text{Te}}^* \propto p_{\text{Te}_2}^{1/2} \propto p_{\text{Cd}}^{-1}$. At high p_{Cd} a different mechanism is expected, D_{Te}^* increasing with p_{Cd} . So far we have obtained one value, $(D_{\text{Te}}^*)_{1000^\circ\text{C}, p_{\text{Cd}}=4\text{atm}} = 3.5 \times 10^{-12} \text{ cm}^2 \text{ sec}^{-1}$.
3. Cd^* tracer self diffusion work by other workers has indicated that D_{Cd}^* is independent of p_{Cd} . Our experiments confirm this only at low p_{Cd} ; at higher p_{Cd} , D_{Cd}^* goes through a weak minimum, then increases $\propto p_{\text{Cd}}^{1/3}$. This indicates diffusion by doubly charged inter-

stitial cadmium, $\text{Cd}_i^{\cdot\cdot}$, the lower constant D_{Cd}^* at low p_{Cd} being due to V_{Cd}'' formed by Frenkel disorder of Cd or by interstitial disorder. (The latter involving Te_i'' and $\text{Cd}_i^{\cdot\cdot}$.)

4. Doping with indium has been found to reduce D_{Cd}^* at high p_{Cd} (by reducing the $\text{Cd}_i^{\cdot\cdot}$ concentration) but increases D_{Cd}^* at low p_{Cd} (by increasing $[V_{\text{Cd}}'']$).
5. High temperature Hall effect measurements in silver doped CdS crystals show that silver has a weak donor action, increasing the electron concentration from 10^{17} to 10^{18} cm^{-3} , at high p_{Cd} , but gives rise to almost intrinsic CdS at low p_{Cd} . Curvature of the Hall constant as $f(p_{\text{Cd}})$ indicates the presence of holes, with $[h^+] > [e^-]$, but still with an n-type sign due to a mobility ratio $v_e / v_h \gg 1$.
The overall behavior indicates amphoteric behavior of Ag, leading to compensated samples at larger concentrations.

Intended Experiments

- I) Investigations of the indium diffusion anomalies by a study with varying indium surface activities, and diffusion times.
- II) Extension of Te^* diffusion at high p_{Cd} to other p_{Cd} and T (owing to the smallness of D_{Te}^* , diffusion times of ≈ 1 month per run are involved).
- III) Extension of Cd^* tracer experiments in indium doped crystals at various p_{Cd} , T and $[\text{In}]$ to defect mobilities and thermodynamic parameters of defect formation.
- IV) Determination of the dominant defect compensating for $\text{In}_{\text{Cd}}^{\cdot}$ at low p_{Cd} (Te_i'' or V_{Cd}'') by accurate measurements of the lattice constant and the density as $f([\text{In}])$.
- V) Carry out high-temperature Hall effect measurements on CdTe-In.
- VI) Determine the solubility of silver in pure and doped CdS, CdTe to see whether oppositely charged Ag species are present.



A Possible Level Scheme for CdTe

Figure 1

e.1. Theoretical Studies of Absorption Mechanisms in IR Window Materials

R. W. Hellwarth

In order to assess the potential performance of various candidate materials for high-power laser windows, we have embarked on calculations of the lower limit placed on the infrared and optical absorption coefficient at "window" wavelengths by various fundamental absorption mechanisms. At frequencies below the electronic band edge, the universal exponential dropoff in absorption (Urbach tail) seems to be fairly well understood.¹ However, at frequencies between 2 and 50 times the fundamental lattice absorption frequency (the lattice absorption tail) the limiting absorption is not understood. It is in this lattice absorption tail where high power operation is currently of most interest and where the measured absorption in candidate materials is generally unacceptably high.

In a perfectly insulating lattice with no vacancies, interstitial ions, or impurities, two mechanisms give rise to the lattice absorption tail: (a) the small anharmonicity in inter-nuclear forces permits the decay of a virtual phonon, created by a photon, into several (n) phonons whose energy adds to that of the absorbed photon; and (b) the (slightly) nonlinear dependence of the electric dipole moment on the lattice coordinates permits the direct decay of a photon into several phonons. Of course vacancies and interstitials always exist at finite temperatures, but our present feeling is that their main effect on the lattice absorption tail appears via altered anharmonicity and nonlinearity parameters in the "anharmonic lattice" and "anharmonic moment" mechanisms of (a) and (b) above. Therefore we have concentrated our efforts in calculating the absorption from these mechanisms for various models. We have not yet found a method of treatment of this absorption which is both tractable and for which the errors can be estimated. Therefore in this report we will simply describe some of the theoretical problems which we are studying.

In principle the anharmonic lattice contributions to absorption can be calculated by standard many-body diagrammatic perturbation. The application of this method to lattice absorption has been reviewed by Cowley.² However this method presents formidable difficulties

when the number n of phonons created by an absorbed photon is much larger than 1:

1) There are an infinite number of types of interaction vertices corresponding to the terms in the infinite Taylor expansion series for the internuclear potentials (in powers of the deviations of their positions from equilibrium). This is in contrast to electrodynamics where there is only one type of vertex to use in diagrams. The coefficients in this series constitute an infinite set of perturbation expansion parameters which, however, can be related to each other arbitrarily for calculational convenience.

2) There is no known algorithm for counting the number of diagrams of a given order in the expansion parameters of the perturbation theory. The number of diagrams for large n becomes very large² and there is no way of knowing when one or more have been omitted. Such inadvertent omissions abound in many-body calculations in the literature and in much less complex situations. However, if, in time, enough people work on a problem, the possible diagrams tend to come to light and be counted. Perhaps diagram-counting algorithms can be discovered to relieve this problem.

3) The coefficient of a certain type of diagram may appear to be smaller than of other types of diagrams and one may therefore be tempted to neglect such terms. However the unknown number of such smaller terms may be large enough to make them important.

4) Unlike in electrodynamics and in electron-hole problems in solid state, "Umklapp" processes are important in the lattice absorption problem. This greatly complicates the integrations over phonon wavevectors which, rather than being constrained to add to zero, need only add to a reciprocal lattice vector. The "Umklapp" processes become more numerous rapidly as n increases.

The approaches we are currently exploring to overcome the difficulties of diagrammatic perturbation theory at large n include:

1) Development of an iterative method for proceeding from terms n to $n + 1$ which in essence automatically constructs all possible diagrams for certain models.

2) Development of space-time perturbation theory to replace the

usual frequency-momentum formulation, thereby omitting specific need to deal with "Umklapp" integrations.

3) The study of the absorption at vibrational harmonics of individual molecules (unit cells), which can be calculated by ordinary Rayleigh-Schrodinger perturbation theory. The possible relation of the discrete -line absorption spectrum of a small number of coupled ions to the spectrum of coupled cells is being sought for high-n.

Typical of our piecemeal results to date is the following observation on the absorption by a molecule at a vibrational harmonic: Here the most complicated terms in the perturbation theory are the most important. I.e. terms that contain the largest number of factors of the total nonlinear potential that contribute for a given n have the largest coefficients, if one assumes for the inter-ionic potentials those derived for the alkali halides with hard-core parameters chosen to give the best fit to the bulk moduli.³

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f. 1 Techniques for Indirect Measurement of Small Absorptive Losses
W.H. Steier

A preliminary version of a calorimetric optical absorption measuring apparatus has been constructed and tested with samples borrowed from AFCRL.

The calorimeter consists of a sample mount with dual thermocouples as temperature sensing elements. A long cylindrical plexiglass shield is used to minimize air currents. A powerful (50 watts per line cw), tunable (grating tuned). CO_2 laser is used as the source. TEM₀₀ mode operation of the laser is maintained and to further reduce the spot-size, a focusing mirror ($R = 3\text{M}$) is used.

Through careful study of the work by other researchers making calorimetric measurements and from the experience gained through our own measurements, we have come to the following conclusions:

1. In making a measurement, it is not absolutely necessary to place the sample in a vacuum. In fact, multiple reflections between the sample and the windows can introduce large errors in the results.
2. It is important to eliminate direct illumination of thermocouples through scattering of the CO_2 laser. This can be accomplished by making sure there is no instantaneous change of thermocouple voltage when the laser is turned on or off.
3. Attachment of thermocouples to the sample must be done with great care. Errors are sometimes introduced through thermal resistance between the sample and the thermocouple. Good agreement between thermocouples placed at opposite sides of the sample indicates that thermocouples are properly attached.
4. During laser illumination, even though different parts of the sample are not at the same temperature, the rate of temperature rise is the same at any point on the sample. Therefore, we are justified to calculate the absorption coefficient from measuring the rate of temperature rise. We believe this is simpler and more accurate than illuminating the sample for a known period of time and then measuring the final equilibrium temperature of the sample.

5. The most time-consuming part of the measurement is in directing the CO_2 beam such that it passes through the center of the sample and that it is perpendicular to the sample faces. A HeNe laser is aligned exactly with the CO_2 laser, consequently, the alignment of the sample with the CO_2 laser beam is achieved very conveniently.
6. Two important factors influencing the accuracy and repeatability of the measurement results are the polish and cleanliness of the sample surfaces and the amplitude stability of the CO_2 laser.

We asked for and received some IR window samples from AFCRL. One CdTe sample (#8A) had an average absorption coefficient, as determined by the AFCRL group of 0.03cm^{-1} . However, in this particular sample, the absorption coefficient is known to be non-uniform across the sample face. Using the apparatus built in our laboratory, we made a series of seven measurements and obtained absorption coefficients of 0.0490, 0.0467, 0.0330, 0.0495, 0.448, 0.0416, 0.0371cm^{-1} . The average absorption coefficient is 0.0431cm^{-1} . One particular difficulty we had was to eliminate certain residual direct illumination of the thermocouple by scattered 10.6μ radiation. That may explain why our measured values are consistently higher than the AFCRL value. To correct this problem we are inserting a diaphragm in the laser cavity to obtain better control of the transverse modes and instead of a focusing lens we are using a focusing mirror to reduce the spot-size of the 10.6μ beam. The wide fluctuations in our measured results can probably be attributed to the non-uniformity of the sample. Further tests will be conducted on a more uniform sample.

g. 1 Characterization of Optical Performance of IR Window Systems

J. H. Marburger, M. Flannery

(1) INTRODUCTION.

The following is a brief description of a computer program for numerically integrating a very general vector Kirchhoff's diffraction integral. The program is capable of calculating the light intensity in space and time for any shape of plane aperture and with an aperture amplitude function that is a general function of time and position. This particular form of the computer program produces contour graphs of the intensity in planes parallel to the aperture plane.

(2) THE DIFFRACTION INTEGRAL.

The basic integral used is an approximation of Kirchhoff's integral with a complex vector amplitude, $A(\xi, \eta, t)$:

$$U(x, y, z, t) = \frac{-ik}{4\pi QR} \left(\frac{z_0}{Q} - \frac{z}{R} \right) e^{ik(R+Q)} \iint A(\xi, \eta, t) e^{ikf(\xi, \eta)} d\xi d\eta \quad (1)$$

The integration is carried out over the aperture in the $z=0$ plane with cartesian coordinates ξ and η . The source and field points, $P_0(x_0, y_0, z_0)$ and $P(x, y, z, t)$, have distances Q and R from the aperture center. Finally the factor $(z_0/Q - z/R)$ is just the difference between the direction cosines of the source and field points with respect to the center of the aperture and a normal in the $+z$ direction. The usual approximation provided in the program gives $f(\xi, \eta)$ with both Fraunhofer and Fresnel diffraction terms:

$$f(\xi, \eta) = (l_0 - 1)\xi + (m_0 - m)\eta + 1/2 \left[\left(\frac{1}{Q} + \frac{1}{R} \right) (\xi^2 + \eta^2) - \frac{(l_0\xi + m_0\eta)^2}{Q} - \frac{(l\xi + m\eta)^2}{R} \right] \quad (2)$$

where,

$$l_o = x_o/Q, \quad m_o = y_o/Q, \quad l = x/R, \quad m = y/R \quad (3)$$

The program uses distances normalized to one half the x-dimension of the integration grid, d . For example, with a circular window, d will be the window radius. We will indicate these normalized variables with a bar; $\bar{R} = R/d$, $\bar{k} = kd$, etc. For calculating the intensity, the program uses the diffraction integral in the form:

$$I(\bar{x}, \bar{y}, \bar{z}, \bar{a}) = I_o \left(\frac{\bar{k}\bar{R}}{4\pi\bar{R}\bar{Q}} (\bar{z}_o/\bar{Q} - \bar{z}/\bar{R}) \right)^2 \left[\left| \iint \text{Re}(a_x) d\bar{\xi} d\bar{\eta} \right|^2 + \right. \\ \left. \left| \iint \text{Im}(a_x) d\bar{\xi} d\bar{\eta} \right|^2 + \left| \iint \text{Re}(a_y) d\bar{\xi} d\bar{\eta} \right|^2 + \left| \iint \text{Im}(a_y) d\bar{\xi} d\bar{\eta} \right|^2 \right] \quad (4)$$

where $\vec{a}(\bar{\xi}, \bar{\eta}, t) e^{i\bar{k}f(\bar{\xi}, \bar{\eta})} = a_x \hat{e}_x + a_y \hat{e}_y$, $I_o = |\vec{A}_{\max}(\bar{\xi}, \bar{\eta}, 0)|^2$ and θ is a normalized time, dependent on the problem solved.

\bar{R}_o is the focal distance and usually $\bar{R}_o = \bar{Q}$, but for problems involving refocusing of the beam \bar{Q} will be a function of time and $\bar{R}_o = \bar{Q}(0)$.

(3) THE INTEGRATOR.

The program uses the simplest possible two dimensional integrator, merely assigning to each integration zone its value at the center. Both the individual integration zones and the grid of all integration points may be varied from square to rectangular. The densities of integration points in the $\bar{\xi}$ and $\bar{\eta}$ -directions are independent so the dimensions of the zones and the grid are relatively independent. For example, this feature is useful if the amplitude function varies much more rapidly in the $\bar{\xi}$ -direction than in the $\bar{\eta}$ -direction, because we can provide a greater density of points in the $\bar{\xi}$ -direction where it varies rapidly, thus more efficiently distributing the integration points.

The integrator evaluates the integral successively, doubling the density of integration points in each dimension for every repetition, and stops when the difference between two successive integrations is smaller than some test value Δ_t :

$$\Delta = (I_n - I_{n+1})/I_{n+1} \leq \Delta_t \quad (6)$$

Experience has shown that for reasonably smooth illumination, convergence is very rapid once $\Delta < 0.1$. Thus the difference between the last integration and the actual value is much less than Δ . Roughly it goes as follows:

$ \Delta = 0.1$	$I = I_{n+1}(1 \pm \Delta/3)$
$= 0.05$	$I = I_{n+1}(1 \pm \Delta/5)$
$= 0.01$	$I = I_{n+1}(1 \pm \Delta/10)$
$= 0.005$	$I = I_{n+1}(1 \pm \Delta/25)$

Thus the final value of Δ produced for a given point is a rather gross upper bound on the error of the integration.

For a specified value of Δ , the integration converges more rapidly for very flat aperture illumination and $\bar{k} \gg 1$, and for field points with high illumination and $\bar{R} \gg 1$. As any of these factors decreases the convergence becomes less rapid.

Finally, the integrator can be set to integrate over the quadrant $\xi \geq 0$, $\eta \geq 0$, the half plane $\eta \geq 0$, or the whole aperture, depending on the symmetry of the aperture and the amplitude function.

(4) THE APERTURE AND ITS BOUNDARY.

The program can accept any shape of plane aperture through several subroutines that determine the location of integration points with respect to the boundary and correct for edge effects when the points are on or near the boundary. The correction merely adjusts the area of the zone, removing the area outside the boundary.

In most cases it is important to provide good edge corrections to avoid a number of pathologies. First, the integrator spreads points uniformly over the integration grid rather than concentrating them near the aperture edges. Thus in cases where the illumination is large at the edges and very smooth in the aperture, the convergence will be limited by edge effects and the integration points inefficiently used unless proper edge corrections are made.

As another example we can consider a rectangular aperture which has one side lying just inside a line of integration points while the opposite side is just outside such a line. This can effectively shift the position of the aperture sideways by about one zone width. While that may be only 1% or less of the aperture width, it moves the focal point an equal amount, which may be several spot diameters. In this case one might end up searching a much larger field area than necessary.

In the case of circular boundaries, if no edge corrections are made, the focal pattern has slight bulges that make the pattern a very rounded square instead of a circle.

Finally it should be noted that time dependent aperture shapes can easily be included in the program.

(5) THE AMPLITUDE FUNCTION.

Through this subroutine we provide the program with any general time dependent vector amplitude function. There are no limitations on the function although functions with rapid intensity or phase changes may require a large number of integration points for proper convergence.

We have made test runs with circular Gaussian beams and checked the results against previous integrations using a one dimensional integrator. Trial runs have also been made for Gaussian beams heating circular isotropic windows, including strain effects, and these calculations agree with a one dimensional program designed specifically for that problem. Finally we have made a few runs on acentric beams with the intensity distribution:

$$I(\bar{r}, \bar{\eta}, \theta) = I_0 \left(\frac{\bar{r} - \bar{r}_0}{\bar{r} - \bar{r}_l} \right) e^{-W((\bar{r} + \bar{r}_0)^2 + \eta^2)} \quad (8)$$

and have included the simple thermal bulging and refractive index modulations due to heating from this beam. Some of these results are included below as an example of the performance of the program.

The time variable is not directly involved in the diffraction integral, but it is provided as a variable in the program so the aperture and amplitude functions may be time dependent. The time variable can be left as the real time or normalized to a characteristic time of the system. It has generally been convenient in simple problems, whose phase aberrations are linear in time, to choose the normalization such that the "time" is the change in the maximum of the phase aberration.

(6) VARIATION IN TIME AND SPACE.

The program is designed to provide a picture of the beam in time and space. To accomplish this the integrator is nested within four do-loops that vary the time, and \bar{z} -dimension and then the \bar{x} and \bar{y} -dimensions. For a given \bar{z} -plane, \bar{x} and \bar{y} are varied so that the intensity is calculated in the center, $(0, 0, \bar{z}, \tau)$, and then at a sequence of points forming successive squares around that central point. This method was chosen so that the power in the plane could be calculated in each successive square. It is possible to provide \bar{x} and \bar{y} with different increments so the region is a rectangle instead of a square.

The output values can be calculated over the regions:

eighth of a plane: $\bar{x} \geq 0, \bar{x} \leq \bar{y}$

quadrant: $\bar{x} \geq 0, \bar{y} \geq 0,$

half plane: $\bar{y} \geq 0,$

or a whole plane.

The output is normalized to any value of the intensity on the \bar{z} -axis, and is printed both numerically and in contour plots of each plane.

There are also a series of checks to prevent calculation of field values in areas where the power or intensity is too small to be of interest.

(7) SAMPLE PROBLEM.

The intensity function of equation (8) was run as an example problem with the parameters $\bar{w}=2, \bar{\epsilon}_0=0.4, \bar{\epsilon}_u=1.0, \bar{\epsilon}_\ell=1.2$ and a circular aperture of radius d . The aperture intensity is plotted in Fig. 1 with its maximum value normalized to unity.

The aberration was taken to be simple thermal bulging and refractive index modulation with no thermal conduction:

$$\begin{aligned}\varphi(\xi, \eta, t) &= k L \left[(n-1) \left(\frac{1}{L} \frac{\partial L}{\partial T} \right) + \frac{\partial n}{\partial T} \right] \frac{\theta t}{\rho_m C_p} I(\xi, \eta) \\ &= \theta I(\bar{\xi}, \bar{\eta}) / L_{\max}\end{aligned}\quad (9)$$

where the last expression is in terms of the normalized variables. If we assume linear polarization of the beam, the amplitude function is:

$$a_x = \left(\frac{I(\bar{\xi}, \bar{\eta})}{I_{\max}} \right)^{\frac{1}{2}} e^{i\varphi(\bar{\xi}, \bar{\eta}, t)}, \quad a_y = 0 \quad (10)$$

For a beam with $\bar{k} = 2.954 \times 10^5$ and $\bar{R}_0 = 2,000$, figures 2-4 show the time development in the focal plane. All the intensity contours have been normalized to the initial focal intensity, $I(0, 0, \bar{R}_0, 0)$, and all variables appearing on the graphs are normalized computer variables.

Taking a specific case we may assume the output is from a CO_2 laser operating at $\lambda = 10.6\mu$, which will make the window radius $d = 0.5$ meters, and the focal distance $R_0 = 1,000$ meters. If we further assume a 1 cm thick KCl output window with a maximum aperture intensity of 100 watts/cm^2 then $\theta = \pi/4$, $\pi/2$ correspond to $t = 2.27$ seconds and $t = 4.54$ seconds.

Notice that the effect of the acentricity of the initial beam is to cause the focal intensity maximum to shift off center and to produce pronounced departures from cylindrical symmetry in the focal region.

During the next period, we plan to use this program, and other programs for cylindrical symmetry, to simulate selected window designs. We have received information from the Parke Mathematical

Laboratories on a program (TEMP 5) for calculating temperature distribution within a window including thermal conduction effects. This program will be included in our simulation code for the evaluation of window designs in the long pulse regime.

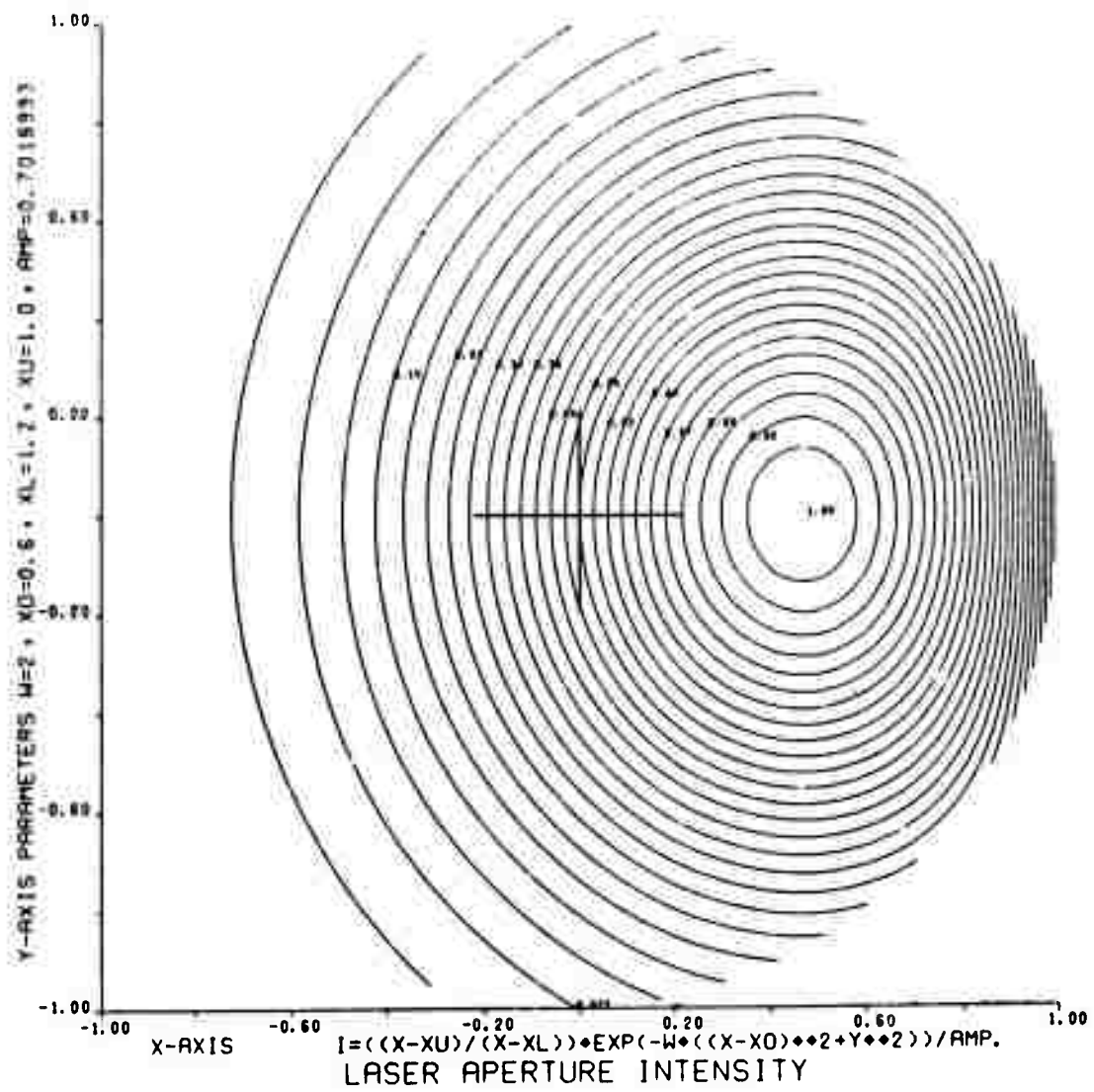


Figure 1

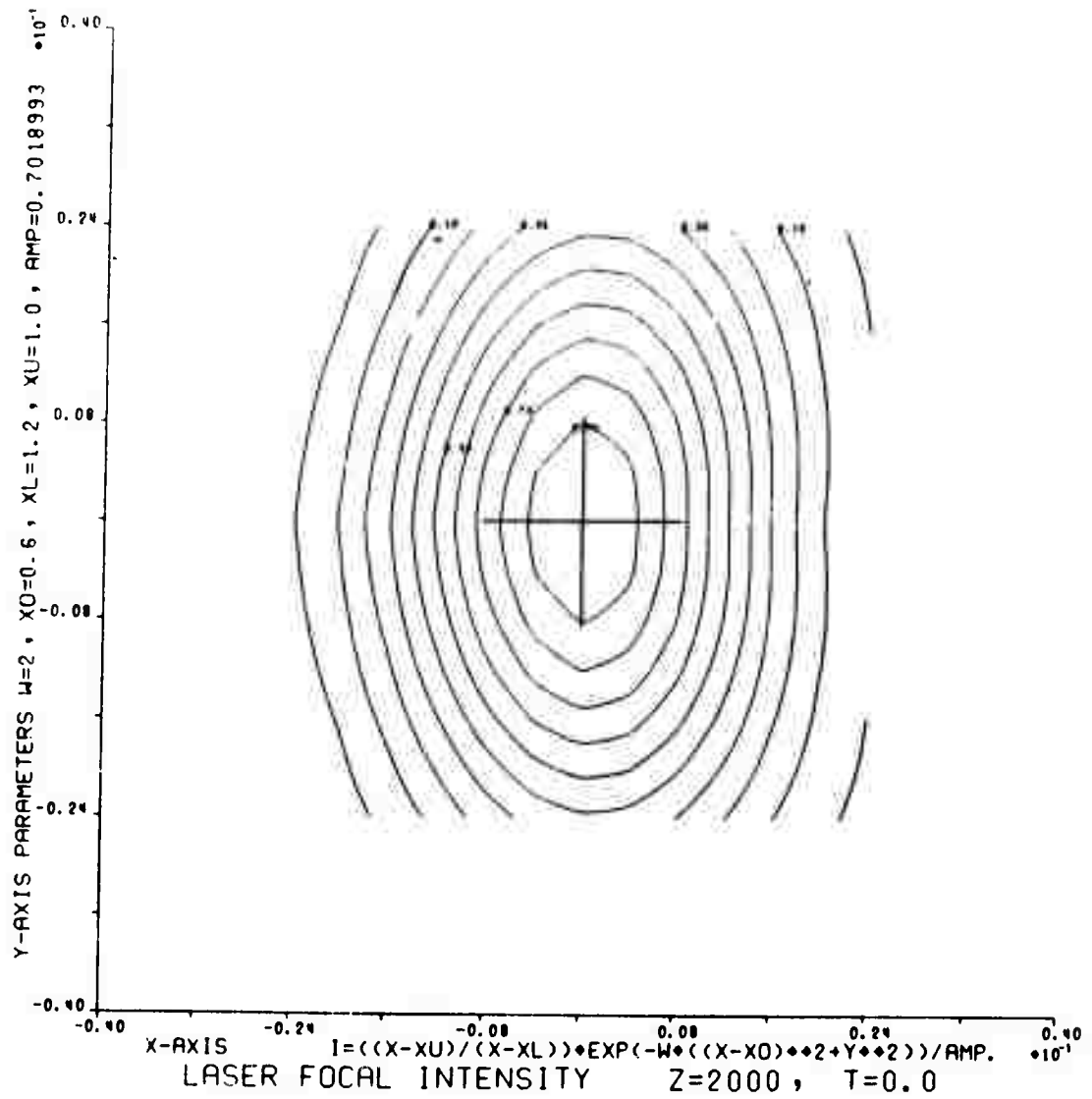


Figure 2

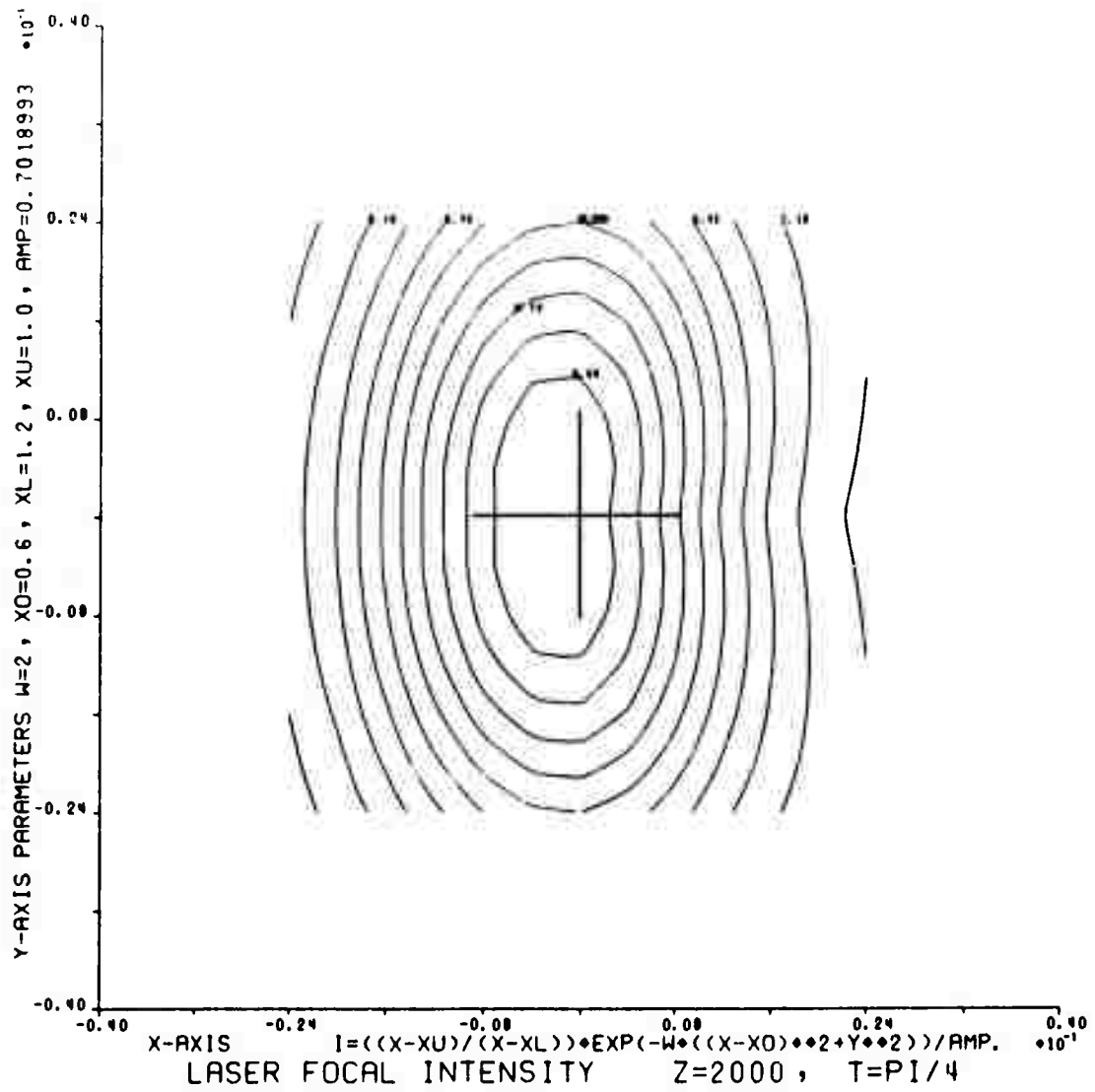


Figure 3

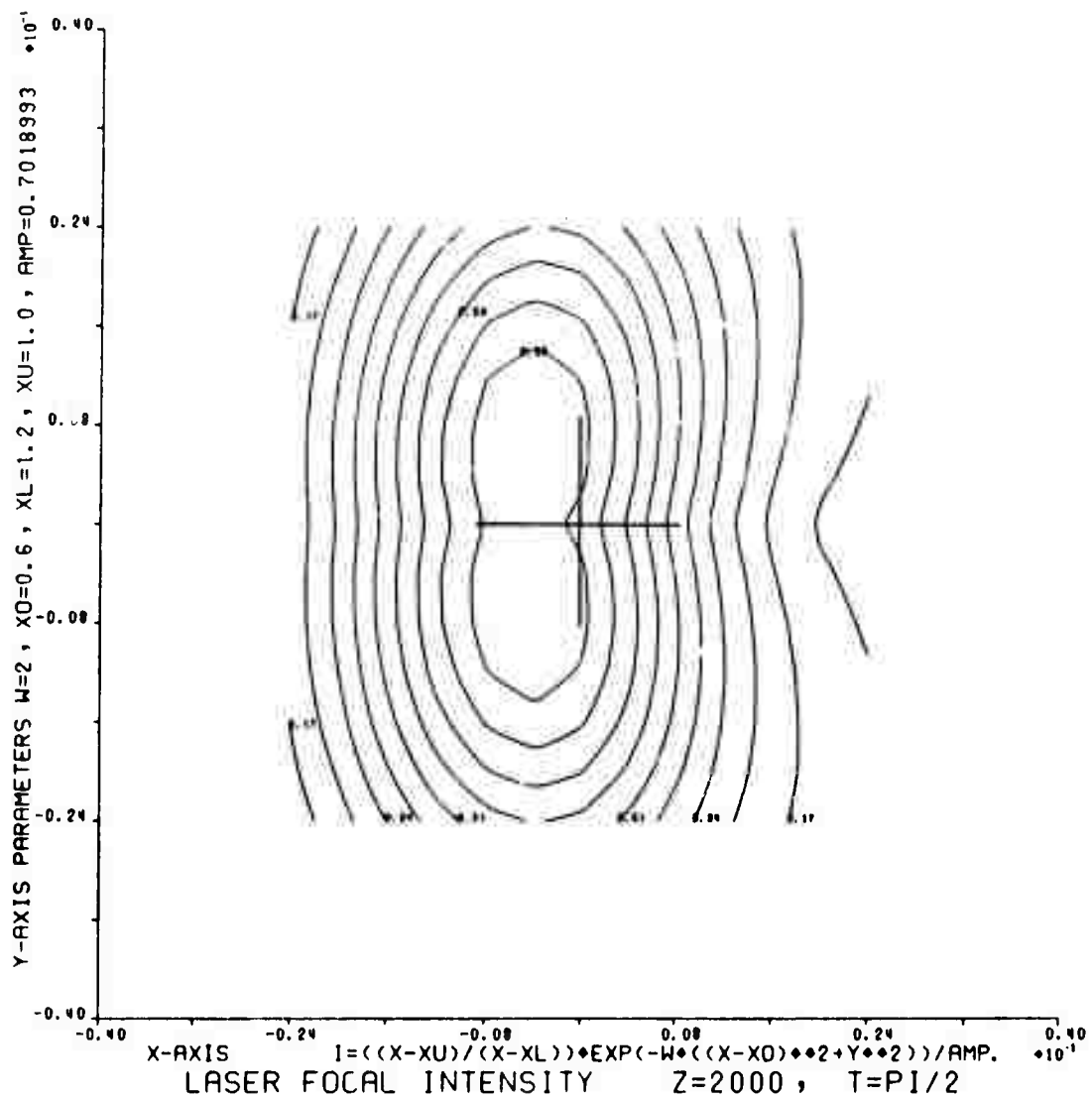


Figure 4

3. DISCUSSION

When the proposal for this contract was prepared, GaAs was regarded as an important candidate for the first IR windows. This, and USC's ~~extensive~~ previous experience with GaAs growth and properties, caused the initial program effort to be strongly oriented toward GaAs. Now that other materials are attracting more attention for window applications, our own attitude toward GaAs has changed somewhat. We feel that in view of the excellent mechanical properties and well-developed technology associated with GaAs, it is extremely important to determine once and for all whether the comparatively high observed absorption at $10.6\text{ }\mu\text{m}$ is intrinsic or a consequence of impurities. The fact that no instance of low absorption has been observed may be related to the fact that virtually all reported measurements have been made on samples grown from the melt, an inherently "dirty" technique. Therefore, we plan to continue those projects whose aim is primarily to grow and to characterize very pure GaAs with controlled impurity content (projects a. 1, a. 3, d. 3). However, we shall broaden the scope of the other projects, where appropriate, to include other promising materials.

During this period, Prof. W. Faust accepted a post at the Naval Research Laboratories, and will no longer participate in the program. The tunable CO₂ laser facility which he constructed, and which we are using for our calorimetry is now being operated by Prof. Steier and Prof. S. P. S. Porto.

Professor Gershenzon, who directs project d. 3, was absent on jury duty during this period. The experimental program for this project is being designed, and a detailed description will be included in the second quarterly report.

During the next quarter, we plan to explore a novel method of probing surface properties under intense $10\text{ }\mu\text{m}$ illumination suggested by Prof. Joel Parks. This method employs phase sensitive detection of microwave surface acoustic waves launched across the illuminated region by an array of transducers deposited on the surface. The technique is sensitive to very small changes of temperature near the surface, and could possibly be used to distinguish between surface and bulk optical absorption. All the technology for these studies is available in-house at USC, and experiments involving acoustic surface waves for other purposes are performed routinely in Prof. K. Lakin's laboratory here.

Several useful discussions were held with AFCRL personnel, particularly Dr. L. Skolnik, regarding our calorimetry apparatus.

4. SUMMARY

A major effort has been initiated at USC to prepare, characterize, and evaluate IR window materials. Initial emphasis is on GaAs because of USC's expertise in this compound, but most studies being undertaken will include other promising window materials.

Preliminary tests of thermal shock resistance of NaCl plates have been conducted. When these were heated above the brittle-ductile phase transition temperature, thermal shock effects were dramatically reduced.

Samples of high purity GaAs were prepared by a closed tube chemical vapor deposition technique. The samples are large enough for optical absorption measurements which are now in progress.

An apparatus for the calorimetric measurement of optical absorption as a function of wavelength in the 9 to 11 μ m region has been constructed and tested. It gave results for a test sample comparable with measurements performed at AFCRL. Design modifications aimed at greater repeatability are now being implemented.

A mask has been fabricated for the deposition of MOS and Schottky barrier structures on semiconductor surfaces for studies of surface and interface IR absorption.

Abnormal concentration profiles of Indium diffusing into CdS and CdTe at 800° C have been observed, indicating unusual behavior of the diffusion coefficient as a function of depth from the surface in these materials. The reason for this behavior is unclear, and will be investigated during the next quarter. Other studies of the kinetics of defects in these materials were also carried out.

Theoretical studies of simple models for multiphonon absorption processes in "transparent" materials have led to a simple criterion for determining the most important terms in perturbation theory which contribute to the absorbance in a certain order. This result is important for the estimation of intrinsic lower limit on absorbance expected for a substance.

A general two-dimensional code has been prepared for the numerical simulation of the optical performance of IR windows. The code differs from previous work in being able to simulate non-cylindrically symmetric situations such as elliptical, acentric, or skewed beams. The code allows the computation of the properties of the optical field as it propagates beyond a thermally distorted window. The effects of

induced birefringence are included. The previously unstudied effect of thermally induced skewing of acentric beams is being examined with this code. (Similar studies have been made elsewhere in the context of atmospheric propagation of intense beams.)